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Title: Nuclear Forensics and Radiochemistry: Fission

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Abstract

Radiochemistry has been used to study fission since it' discovery. Radiochemical methods are used to determine cumulative mass yields. These measurements have led to the two-mode fission hypothesis to model the neutron energy dependence of fission product yields. Fission product yields can be used for the nuclear forensics of nuclear explosions. The mass yield curve depends on both the fuel and the neutron spectrum of a device. Recent studies have shown that the nuclear structure of the compound nucleus can affect the mass yield distribution

Nuclear Forensics and Radiochemistry: Fission

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LA-UR-17-xxxx

Outline of Lecture

- **In the beginning: The discovery of fission**
- **Forensics using fission products**
  - What can be learned from fission products
  - Definitions
    - R-values
    - Q-values
  - Fission bases
  - K-factors and fission chambers
  - Limitations
- **The neutron energy dependence of the mass yield distribution**
  - The two mode fission hypothesis
- **The influence of nuclear structure on the mass yield distribution**
- **Summary**

In the Beginning: Discoveries Set the Stage

- Mass spectrometry was invented 1897.
- Soddy postulates the existence of isotopes 1913.
- Thompson has first evidence of stable isotopes 1913.
- Einstein publishes the theory of special relativity 1916.
- These led to the nuclear curve of binding energy.
- Chadwick discovers the neutron in 1932. He used a source of 2 mCi of Polonium-210 mixed with beryllium.
- In 1934 Enrico Fermi began inducing radioactivity with a polonium-beryllium source.
- The stage was set

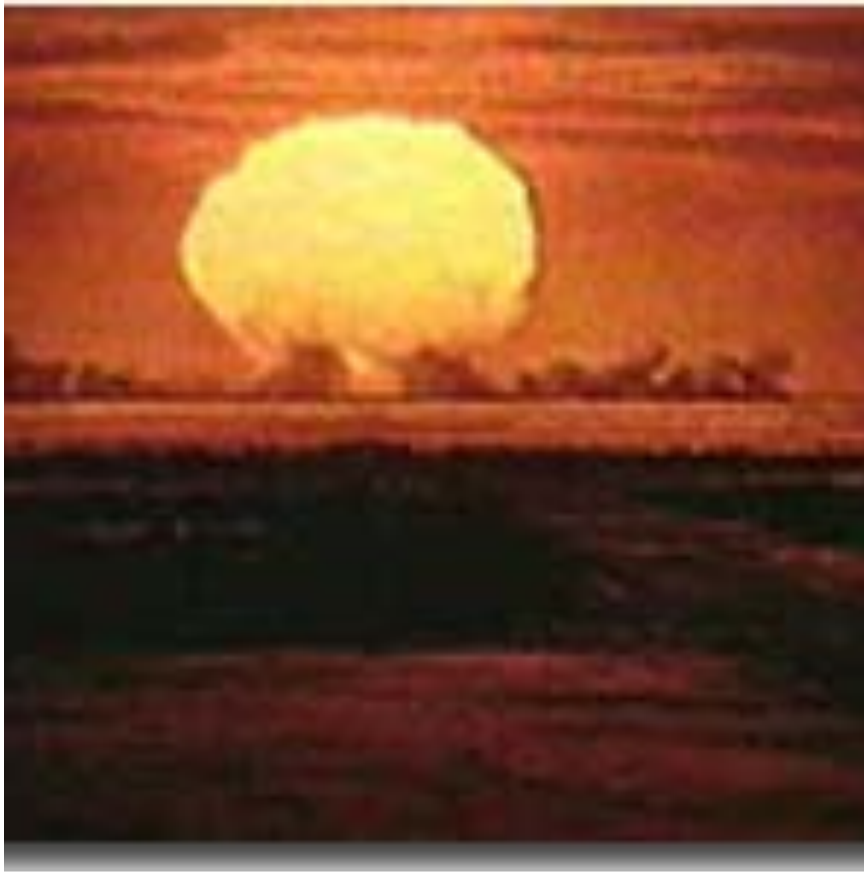
In the Beginning: The Discovery of Fission, 1938




- Otto Hahn, Fritz Strassmann, with Lisa Meitner attempt to make new isotopes by irradiating uranium with neutrons from a radium-beryllium source.
- They had hope to make a new radium isotope.
- After dissolving the uranium barium was added as a carrier. After separating barium, radium was to be extracted by fractional crystallization of $RaCl_2$ (solubility 245 g/ L) from $BaCl_2$ (solubility 358 g/L)
- The induced radioactivity never separated from barium.
- The conclusion was that a radioactive isotope of barium was formed, i.e., fission.
- Frisch and Meitner postulated fission.

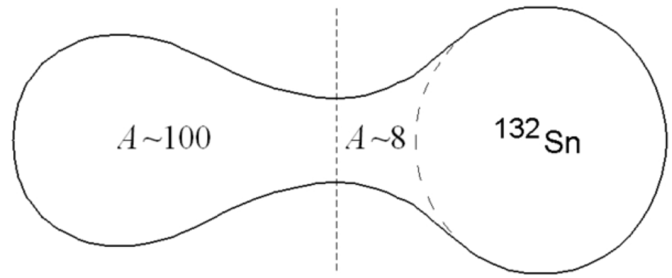
The Nuclear Age Begins

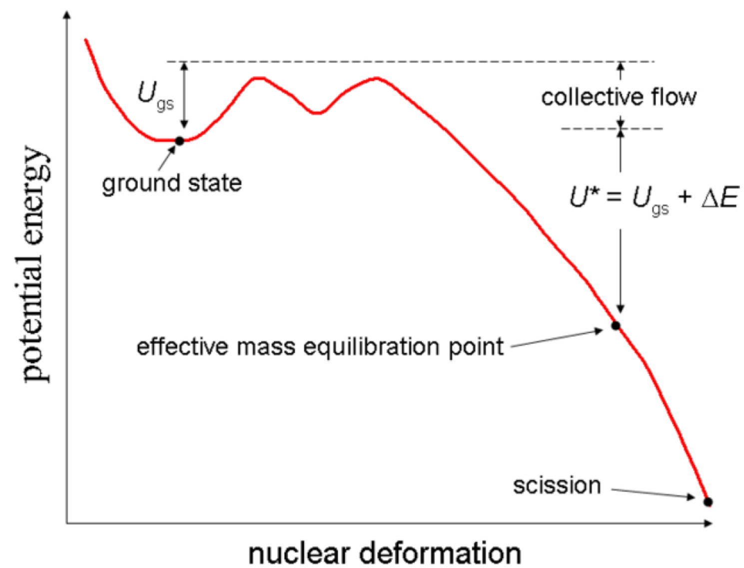
- In 1939 realizing the enormous energy release from fission Leo Szilard and Eugene Wigner composed a letter to President Roosevelt.
- Preparations began in 1941, and the Manhattan project was started in 1942.
- 16 July, 1945 The Trinity test was detonated on the White Sands missile range near Alamogordo New Mexico.
- World war 2 ended Sept. 2, 1945



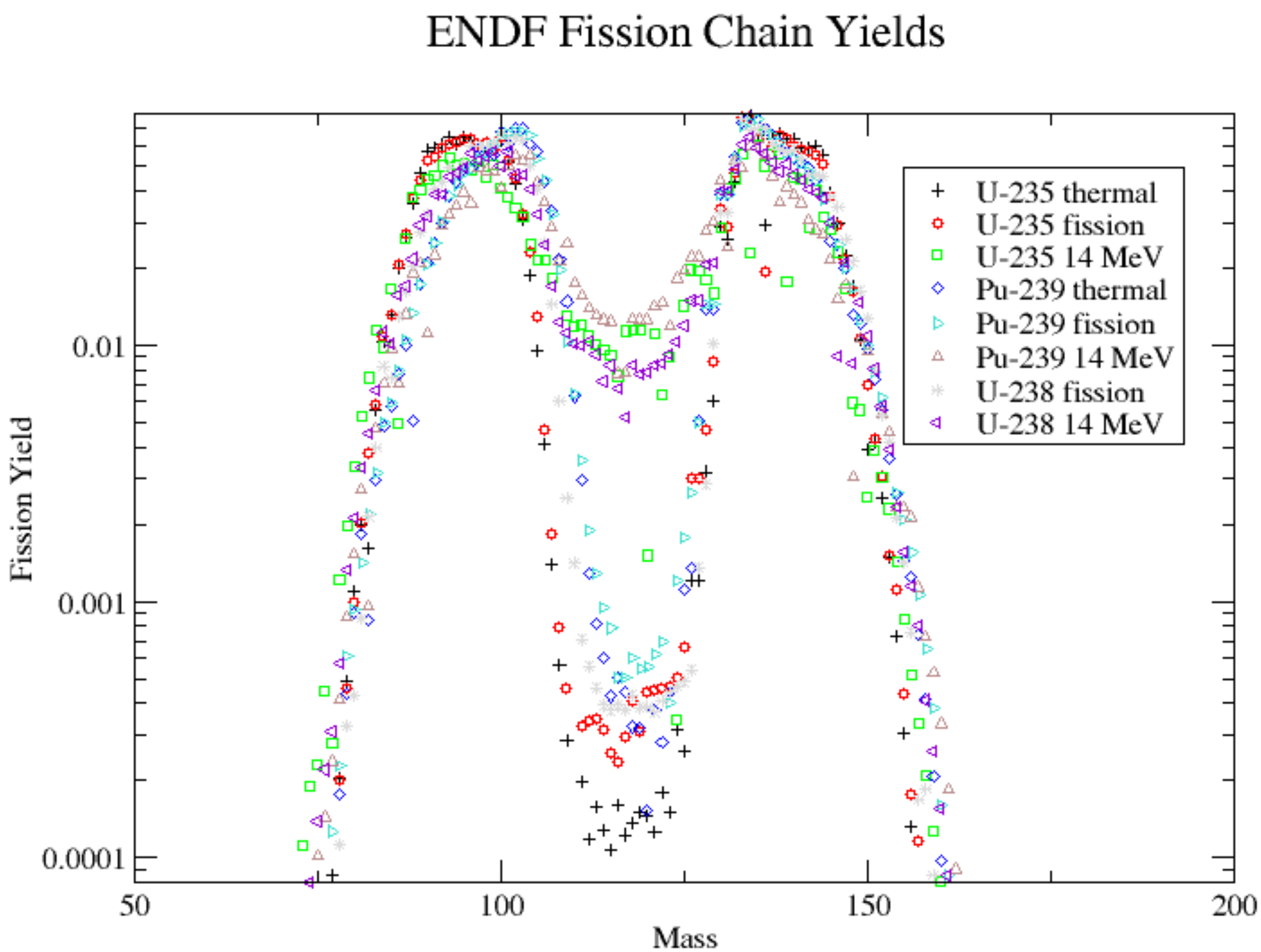
Fission Products: Scientific Background

- The mass yield curve depends on the mass of the fissioning nucleus and the neutron energy.
- Differences in the measured yields allow the determination of the identity of fissile materials and the neutron energy distribution.
- The asymmetry in the mass yield curve is due to lowered potential energy from the ^{132}Sn core in heavy fragment.
- Excess neutrons from the neck cause a broadening and shift to about mass 140.





ENDF Fission Yields



Scientific Background: R-values

- R-values are a ratio of activity ratios.
- The denominator is a reference. Namely the thermal neutron induced fission of U-235.
- The advantage of this ratio is that most systematic errors are cancelled. For example, detector efficiency, branching ratios, etc.
- R-values can be used to determine splits between fuel and neutron energy.

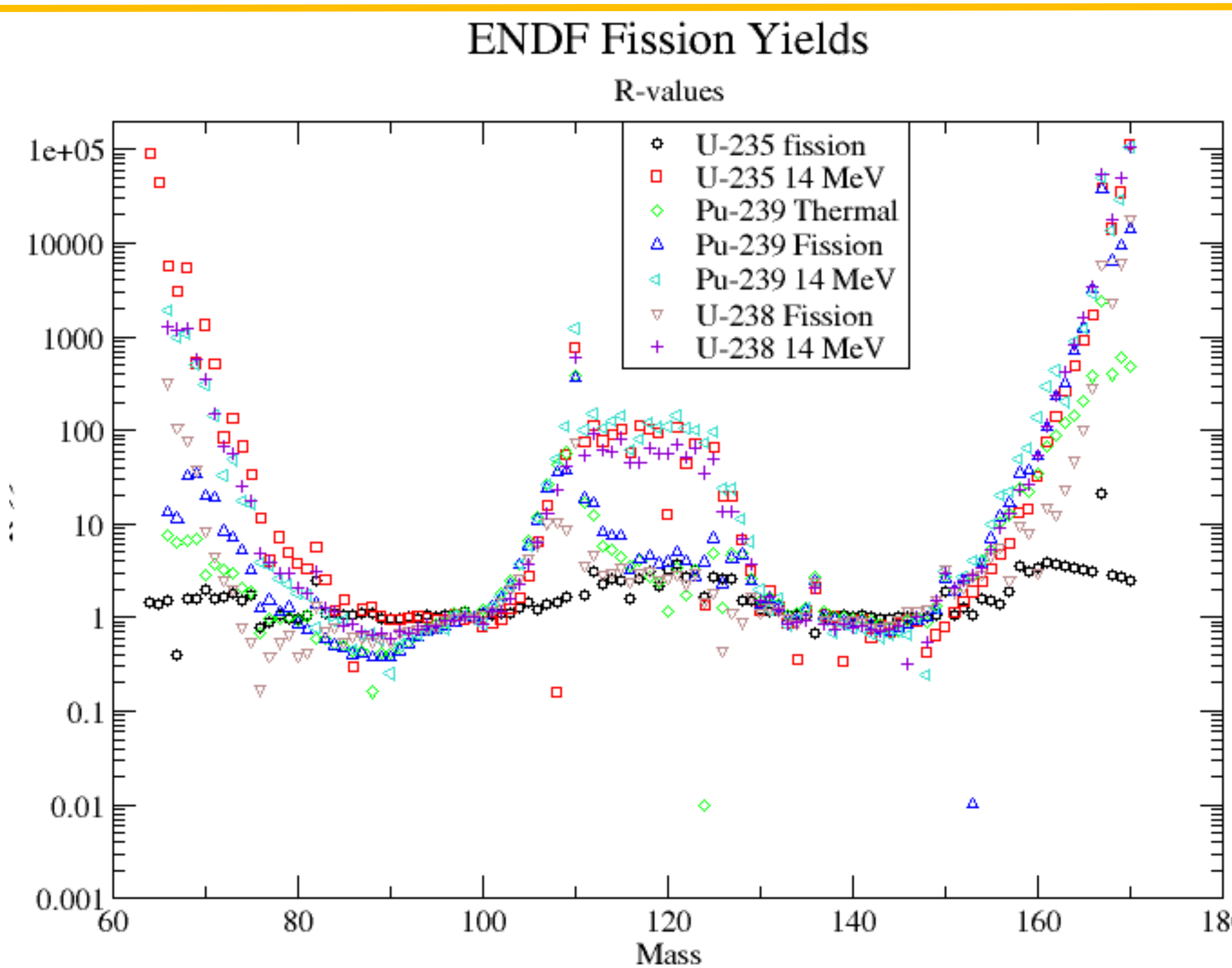
$$R^{x}_{i,99} = \frac{[A^{x}_{i} / A^{x}_{99}]}{\left[A^{U235,T}_{i} / A^{U235,T}_{99}\right]}$$

$$R^{x}_{i,99} = \frac{\left[\varepsilon_i f_i \lambda_i Y^{x}_{i} N^{x}_{fissions} / \varepsilon_{99} f_{99} \lambda_{99} Y^{x}_{99} N^{x}_{fissions}\right]}{\left[\varepsilon_i f_i \lambda_i Y^{U235,T}_{i} N^{U235,T}_{fissions} / \varepsilon_{99} f_{99} \lambda_{99} Y^{U235,T}_{99} N^{U235,T}_{fissions}\right]}$$

$$R^{x}_{i,99} = \frac{\left[Y^{x}_{i} / Y^{x}_{99}\right]}{\left[Y^{U235,T}_{i} / Y^{U235,T}_{99}\right]}$$

Efficiencies depend on detector, sample preparation, geometry. Gamma ray abundances also depend on literature values, internal conversion coefficients, etc.

ENDF R-Values



LANL's Approach Uses R-Values, Q-Values: Ratios Allow Us to Minimize Systematic Errors

$$Y_{147}^{Pu,fast} = Y_{147}^{235,th*} \cdot \overbrace{\frac{\left[Y_{147}^{Pu,fast} / Y_{99}^{Pu,fast}\right]}{\left[Y_{147}^{235,th} / Y_{99}^{235,th}\right]}}^{R_{147}^{Pu,fast}} \cdot \overbrace{\frac{Y_{99}^{Pu,fast}}{Y_{99}^{235,th}}}^{Q_{99}^{Pu,fast}}$$

Often accurately measured
(many thermal reactor experiments)

R: Precise "Lab independent" Measurements – ratio of ratios.

Cancellation of systematic errors In counting FPs (99 and 147)

Cancellation of # of fissions for A given neutron environment

Q: "lab dependent" and difficult absolute measurement (K-factors from fission chamber experiments) needed for both numerator and denominator,

BUT, cancellation systematic errors in counting FPs (99). Errors in the # of fissions remain.

Forensics from R-values

- **We want to be able to express R-values measured from debris of an unknown detonation in terms of end-point R-values and Q-values.**
- **End-points refer to possible fuels (*j*), i.e., U-235, Pu239, and U-238, and neutron energy groups (*k*).**
- **We consider two energy groups, fission spectrum (slow), and 14.1 MeV (fast).**
- **By measuring a suite of fission products (*i*). We determine by least squares fitting the coefficients *a* (the number of fission from each fuel and energy group).**

$$Q_i^{unkown} = \left(\frac{Y_i^{unkown}}{Y_i^{235,th}}\right) = \sum_{j=a,k=s}^{c,f} a_{j,k} R_i^{j,k} Q_{99}^{j,k}$$

$$Q_{99}^{unkown} = \left(\frac{Y_{99}^{unkown}}{Y_{99}^{235,th}}\right) = \sum_{j=a,k=s}^{c,f} a_{j,k} Q_{99}^{j,k}$$

$$R_i^{unkown} = \left(\frac{Q_i^{unkown}}{Q_{99}^{unknown}}\right) = \frac{\sum_{j=a,k=s}^{c,f} a_{j,k} R_i^{j,k} Q_{99}^{j,k}}{\sum_{j=a,k=s}^{c,f} a_{j,k} Q_{99}^{j,k}}$$

Fission Measurements: K-Factor

- **The K-factor is a constant that converts the count rate of a fission product in a sample to the number of fissions.**
- **Historically beta counters were used to measure the radioactive fission products. There are two advantages to beta counting: 1) high counter efficiency, 2) branching equals 1.0. Beta counting requires the same detector be used every time, the same sample thickness and geometry.**
- **K-factors must be determined by fission chamber experiments.**

$$n_{fissions}^{x} = K_i^x A_i^x$$

$$K_i^x = \frac{n_{fissions}^{x}}{\varepsilon_i f_i \lambda_i Y_i^x}$$

Beta requires precise sample preparation. It is not always recognized that changes in sample preparation can also affect gamma ray detection efficiency.

How did we measure endpoint values?

- Thin deposits of fissile material on a substrate, e.g., platinum, surround a thick foil inside a double ionization chamber.
- The chamber is placed in the neutron flux from a bare critical assembly, i.e., Godiva or Jezebel.
- The number of fissions in the thin foils are registered as counts from the gas proportional chambers.
- The efficiency for a thin foil is nearly 100%.
- The number of fissions in the thick foil is scaled to the thin foils by mass.
- The Q-values endpoints are simply the the ratio of the activity per fission in non-thermal flux over the activity per fission of the same isotope from U-235 in a thermal flux.

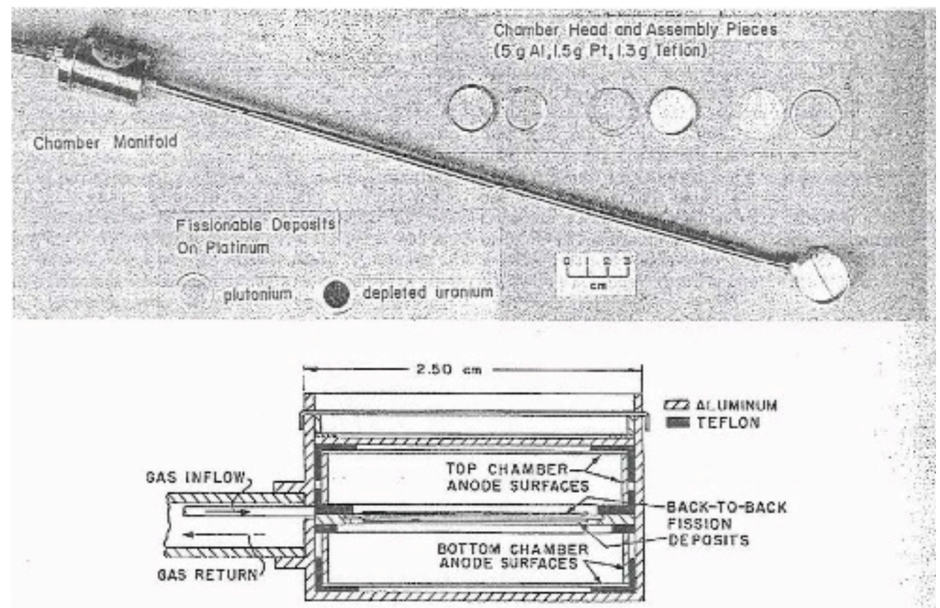
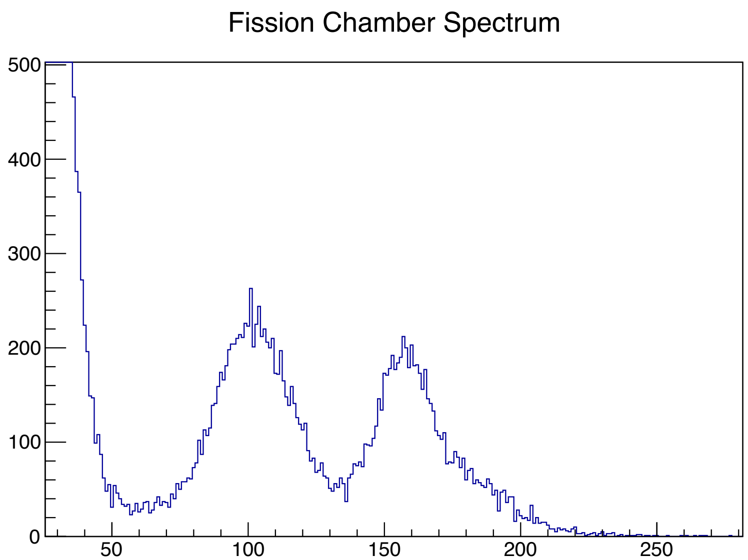


Fig. 1. The drawing shows an enlarged cross-sectional view of the dual chamber head, which is seen on the right in the photograph. The anode connections (+150 V) are made by single-strand Teflon-coated wires running through the stem to each cylindrical anode surface. The top chamber pieces are removable to allow easy exchange of the fissionable deposits.

Fission Chamber Irradiated in the Flattop Critical Assembly



Limitations of Fission Products

- The neutron energy spectrum is treated as only a two group spectrum, namely fission spectrum plus 14.1 MeV.
- The fission spectrum is assumed to be Godiva or Jezebel leakage spectra.
- The end-point R-values are based on few measurements.
- Evaluations of fission yields suggest the need for further measurements for Nd-147 an important reference fission product.
- Fractionation is a problem because of volatility differences between the elements.

The Two Mode Fission Hypothesis

- The 2 fission mode hypothesis assumes that the mass yield curve consists of an asymmetric distribution and a symmetric distribution.
- The hypothesis is that the magnitude of the asymmetric distribution is proportional to the yield of ^{99}Mo.
- The magnitude of the symmetric distribution is proportional to the yield of ^{111}Ag.
- By combine the relationships between yields and R-Values. Then the R-Value of any isotope for a given experiment has a linear relation to R-111.

$$Y_{ji} = a_i Y_{j,111} + b_i Y_{j,99}$$

$$Y_{j,99} = Y_{T,99} Q_{j,99}$$

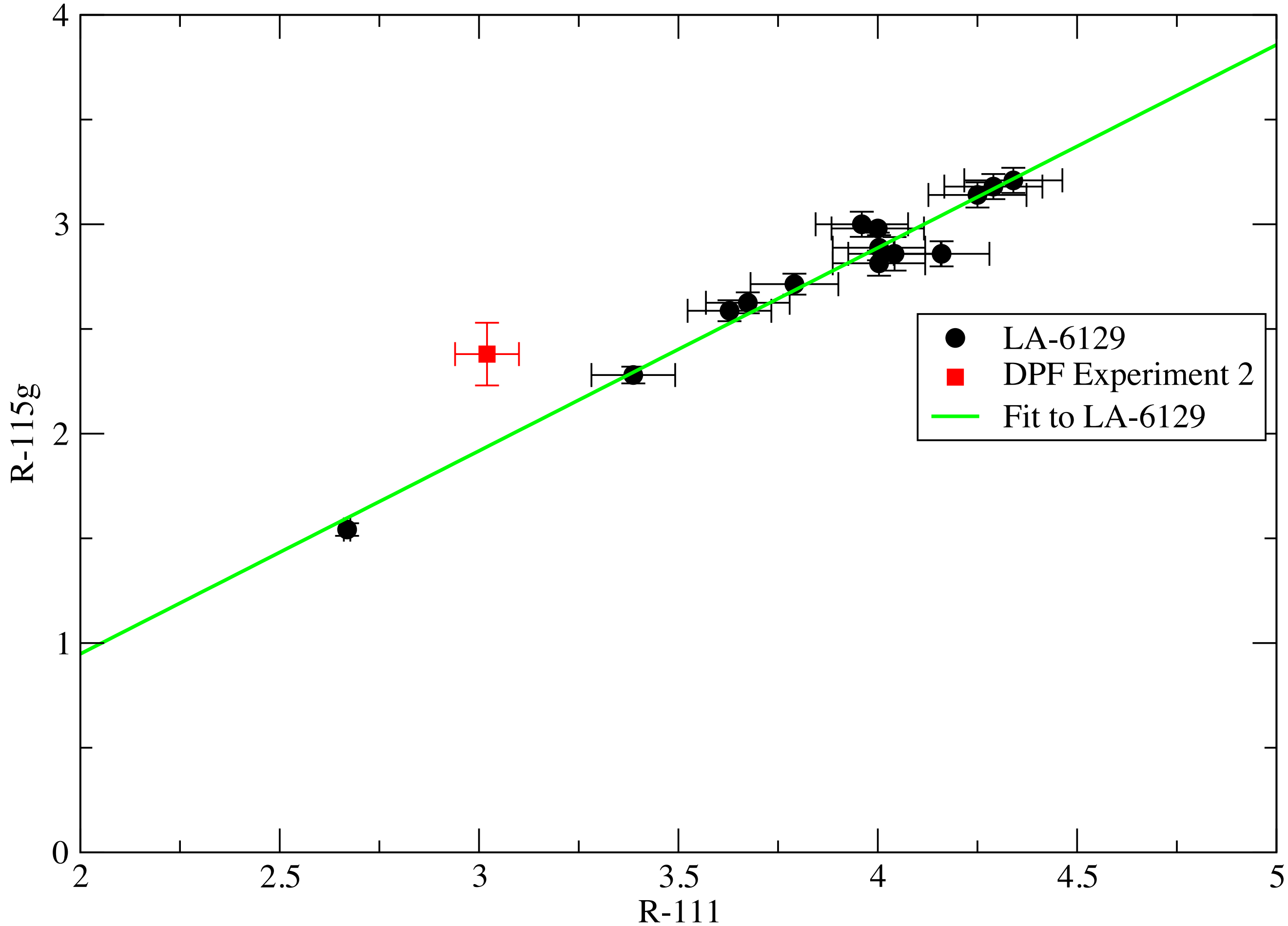
$$Y_{j,111} = Y_{T,111} R_{j,111} Q_{j,99}$$

and

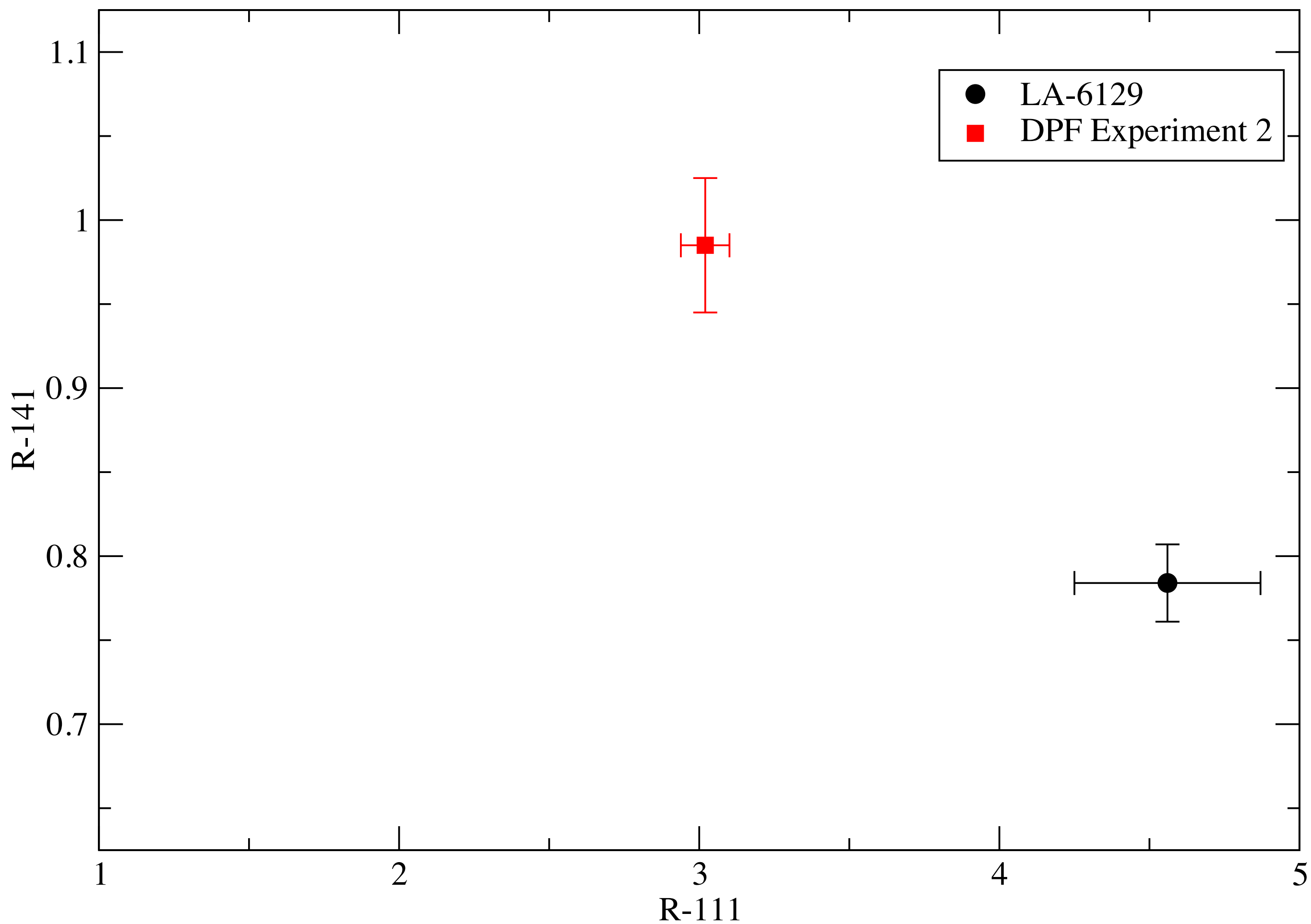
$$Y_{ji} = Y_{T,i} R_{ji} Q_{j,99}$$

$$R_{ji} = a_i \frac{Y_{T,111}}{Y_{T,i}} R_{j,111} + \frac{Y_{T,99}}{Y_{T,i}} b_i$$

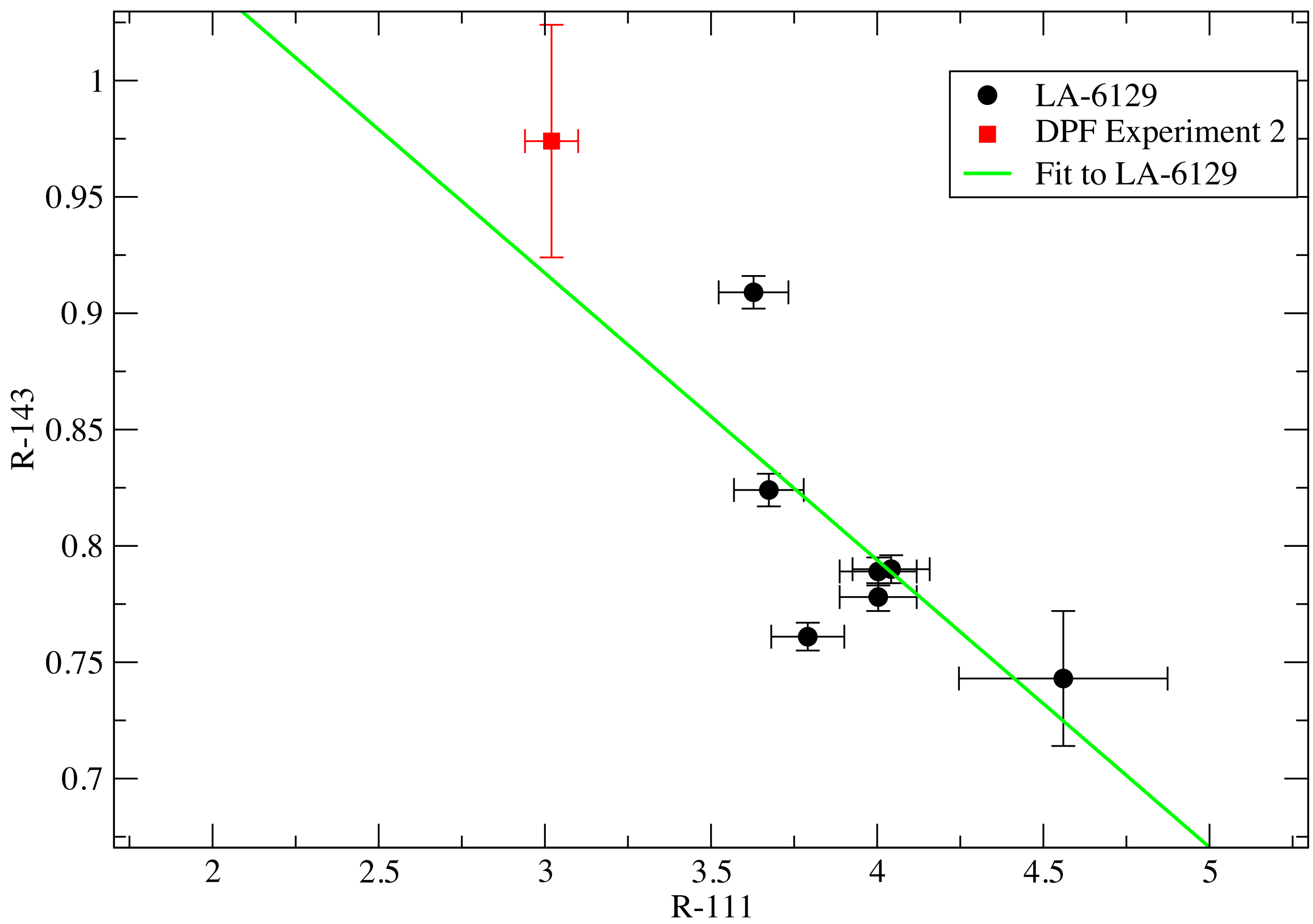
Cd-115g R-values for U-238



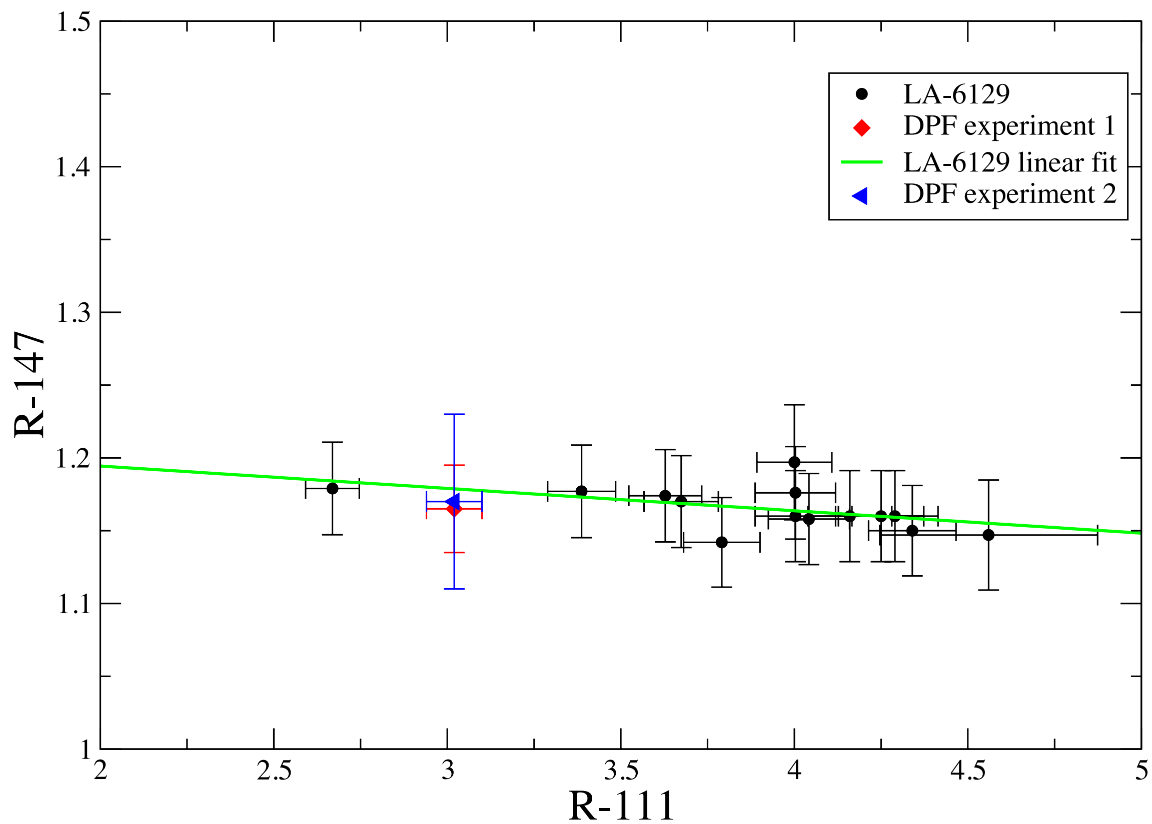
Ce-141 R-values for U-238



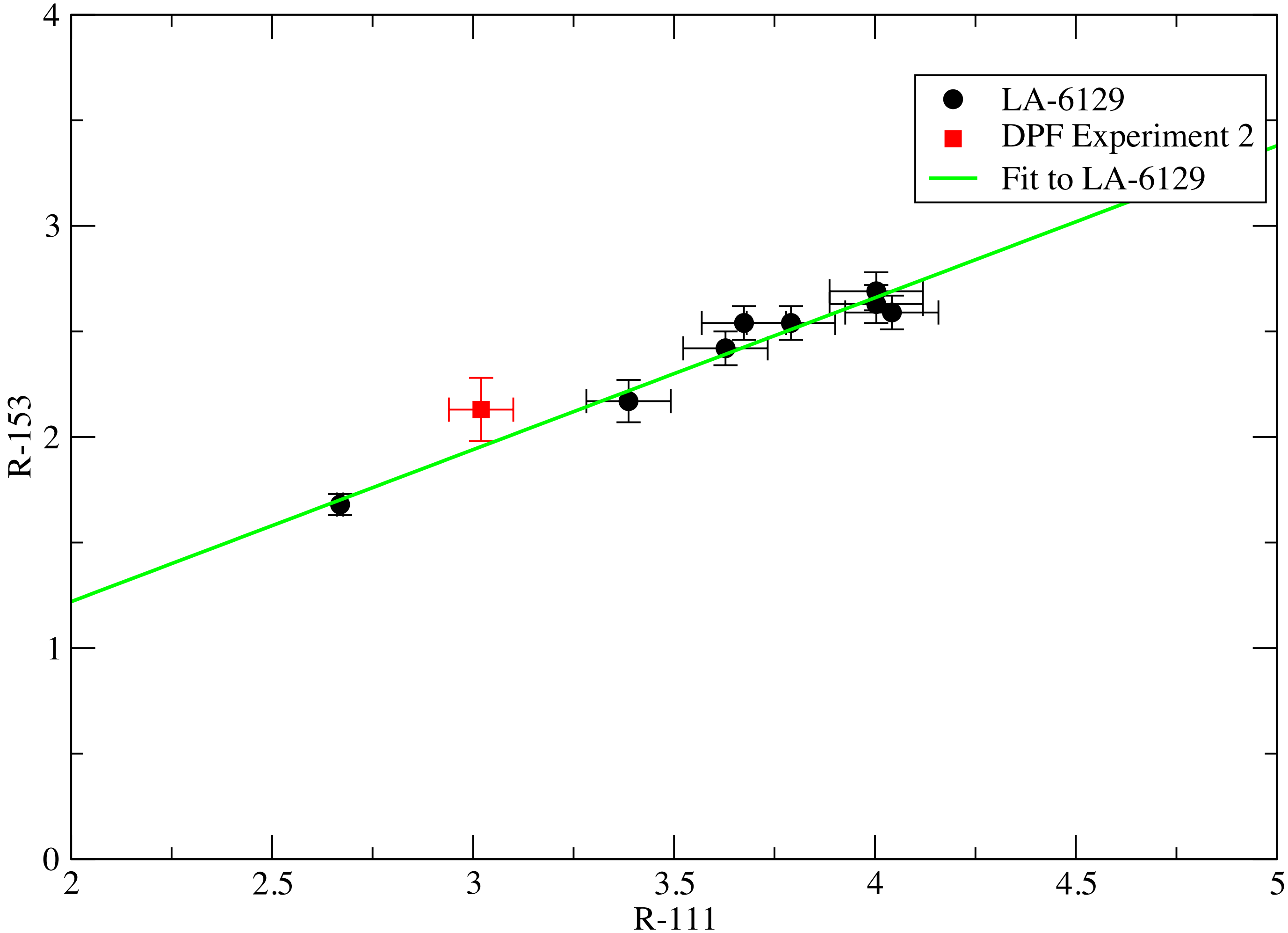
Ce-143 R-values for U-238



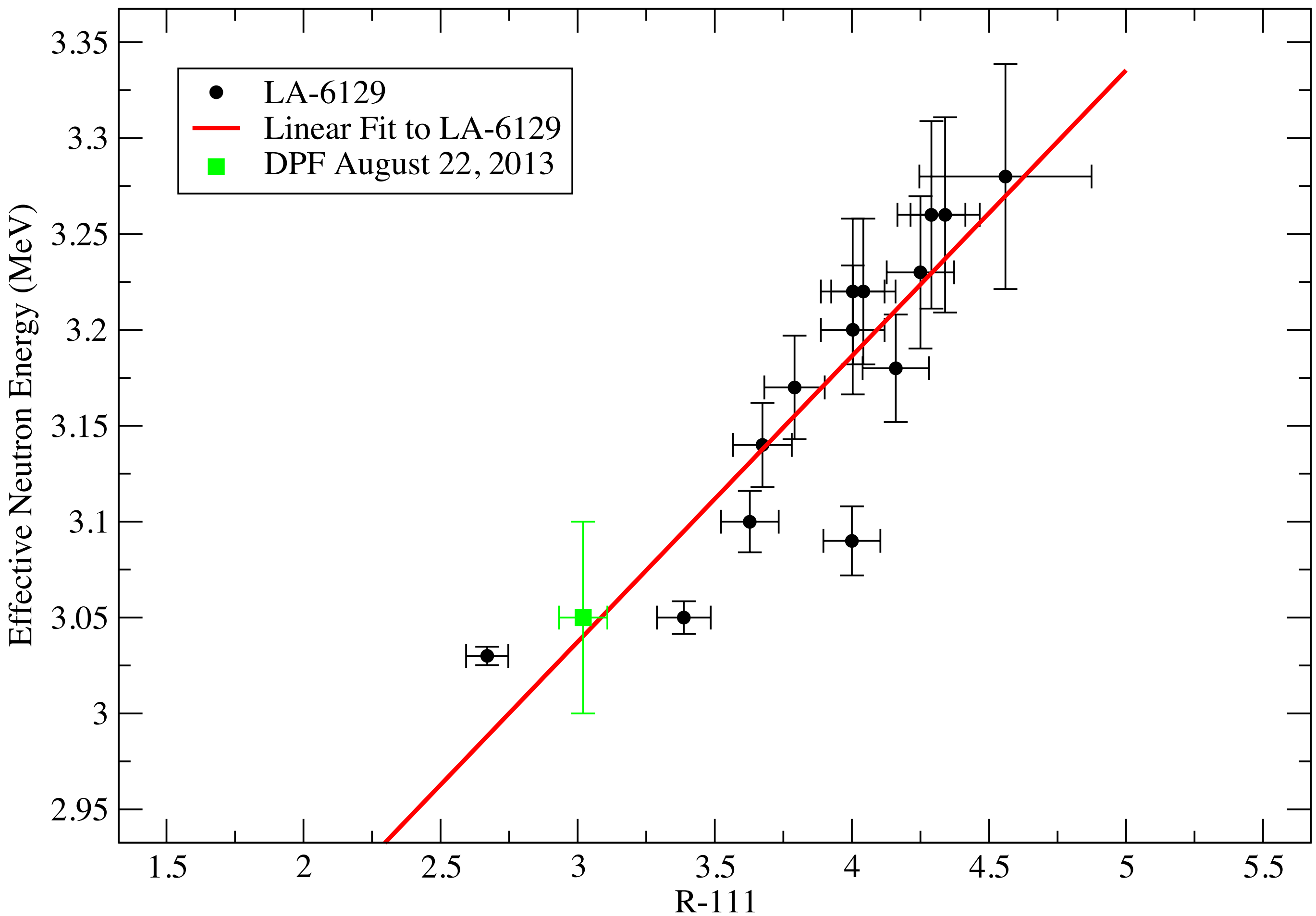
Nd-147 R-values for U-238



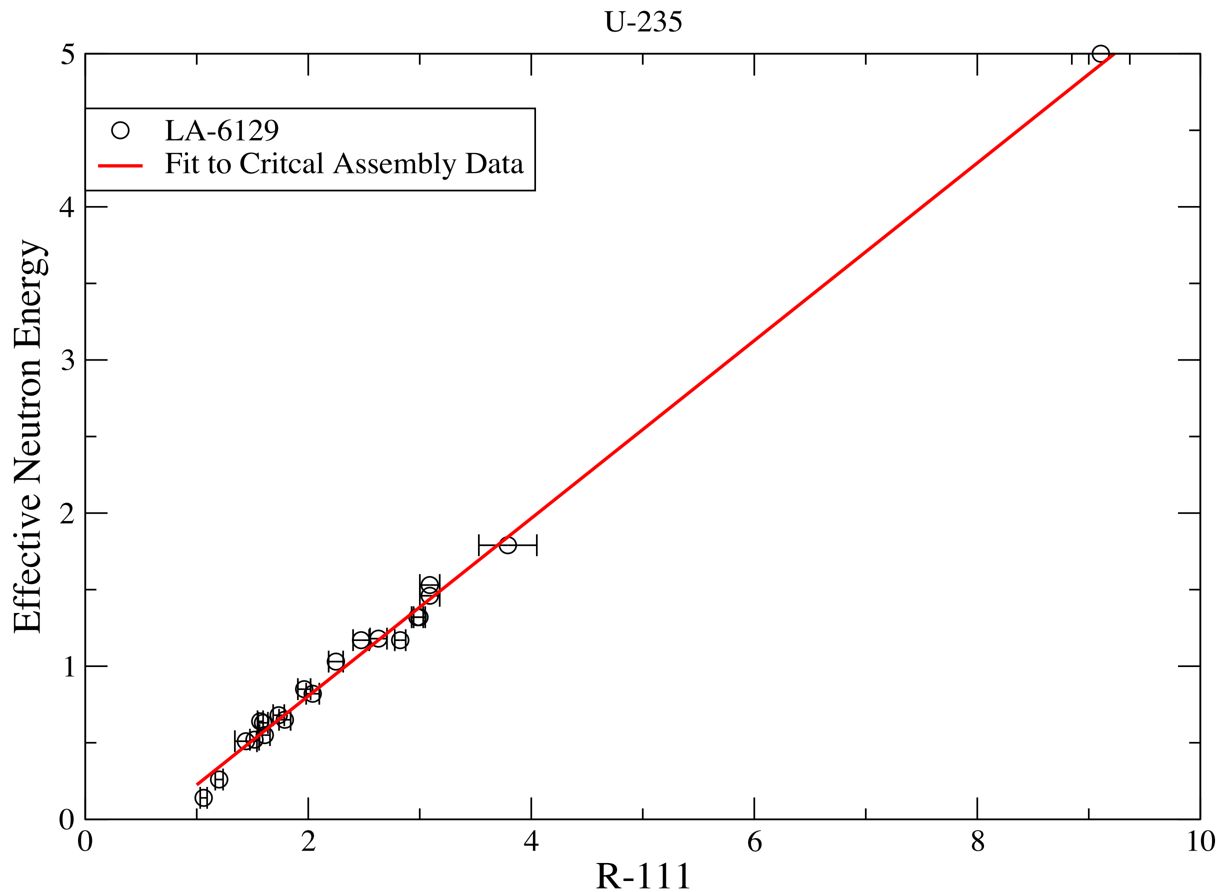
Sm-153 R-value



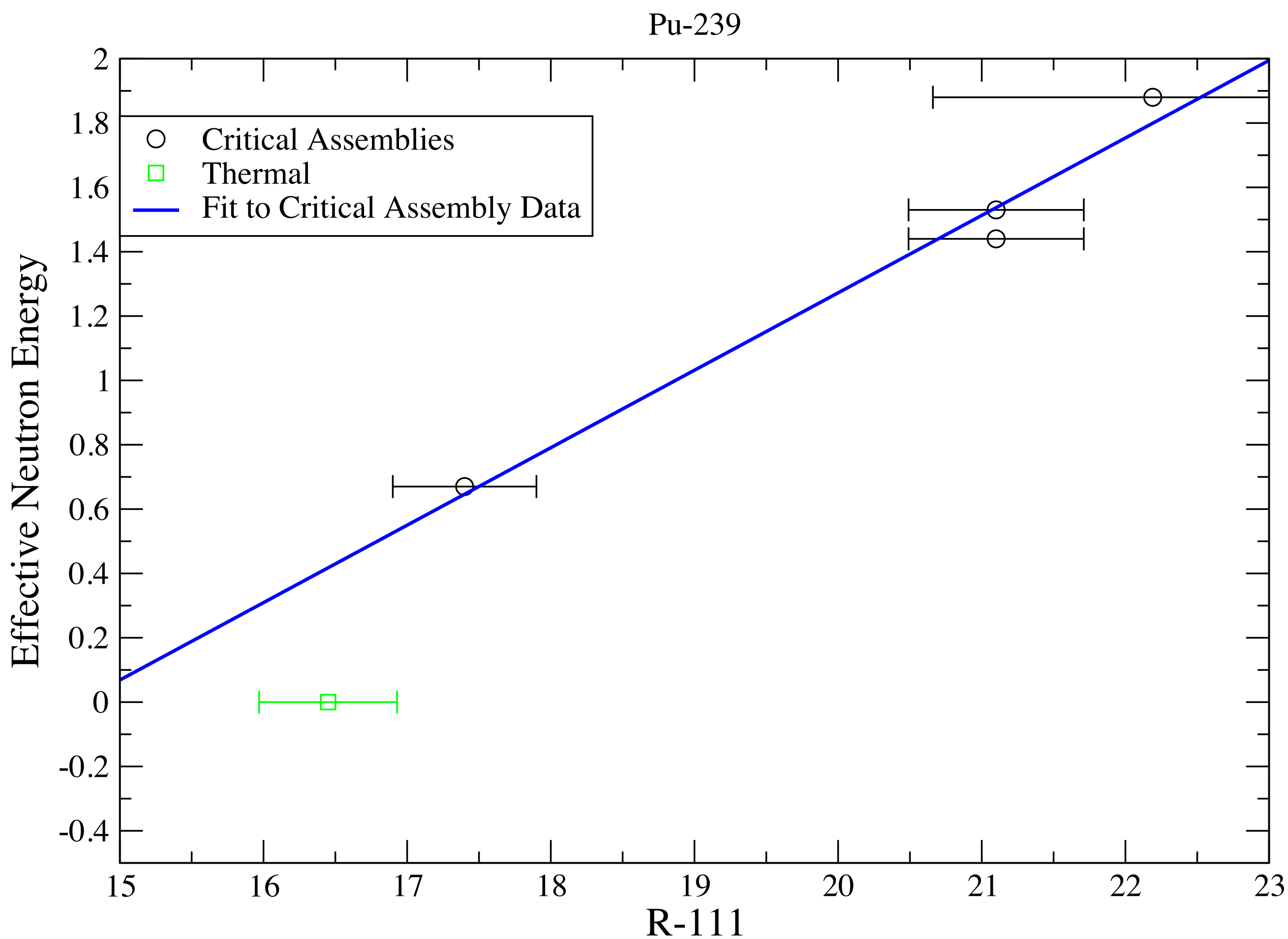
Neutron Energy Dependence of R-111 for ^{238}U



Effective Neutron Energy for Critical Assemblies

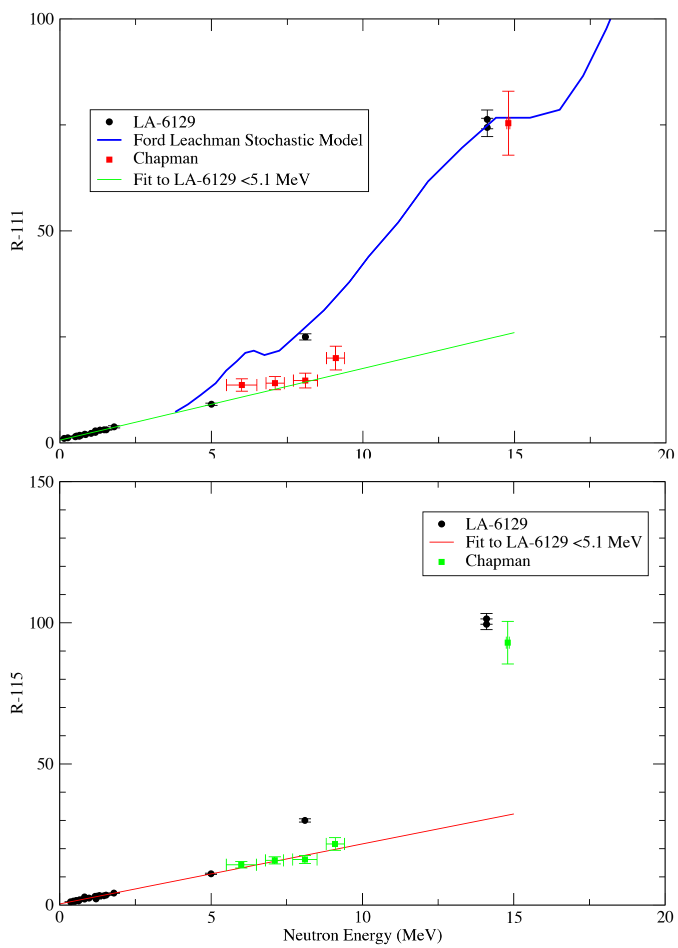


Effective Neutron Energy for Critical Assemblies

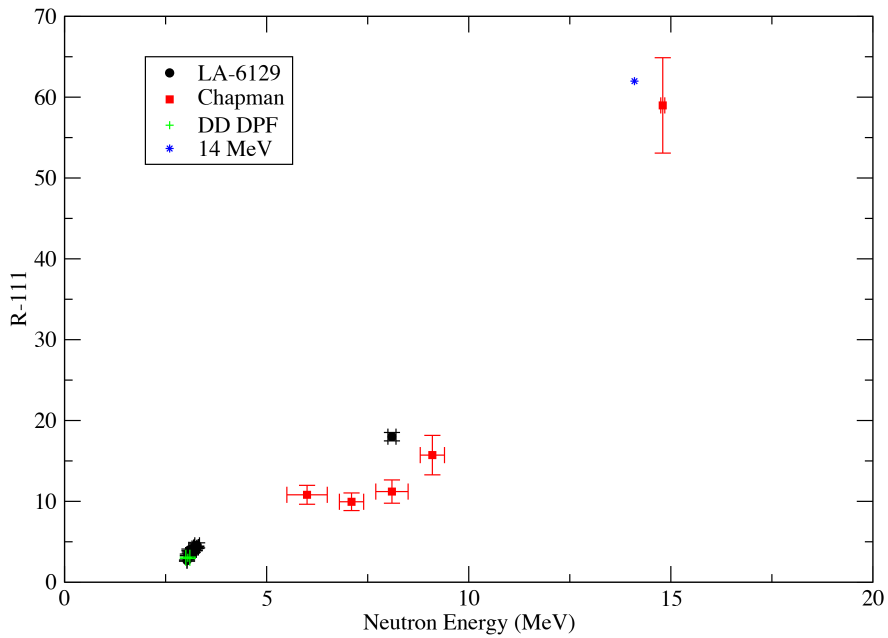


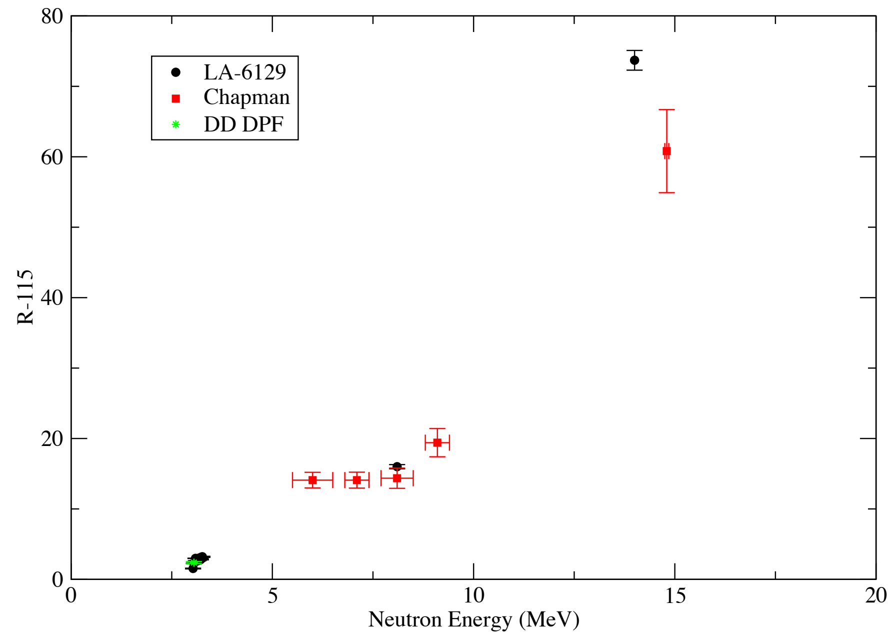
Fission yields from Symmetric Fission of U-235

- **Fission yields are used in post-detonation forensics to identify the nuclear fuels and neutron energy spectrum.**
  - R-values are a double ratio that is approximately proportional to the yield.
  - The figures show the neutron energy dependence of R-111(Ag-111) and R-115 (Cd-115g).
  - The blue curve in the upper figure is the stochastic model of Ford and Leachman, Phys. Rev. 137, 826(1965)
  - The colored data points were derived from the fission yields reported in the Thesis of Terry Chapman, AFIT (1978).
    - There appears to be a systematic error introduced by combining Chapman's yields with England and Rider.
    - The shape generally agrees with Ford and Leachman.



Fission Yields from Symmetric Fission of U-238





- **The data is sparse in the region around the onset of second chance fission.**
  - More measurements are needed to develop a consistent basis and enable the development of reliable model for the two-mode fission hypothesis.

Resonances Affect the Mass Yield Distribution

- In 1956 John Wheeler suggested that the fission mass yield distribution in neutron resonances should differ from thermal neutron induced fission, Physica 22, 1103(1956).
- Shortly after the Los Alamos verified this hypothesis by irradiating ^{235}U foils in the Fermi water boiler reactor with cadmium shields.
- R-111 was 0.9 with a cadmium shield and R-115g was 0.86.
- This motivated the “Wheel Experiment.”

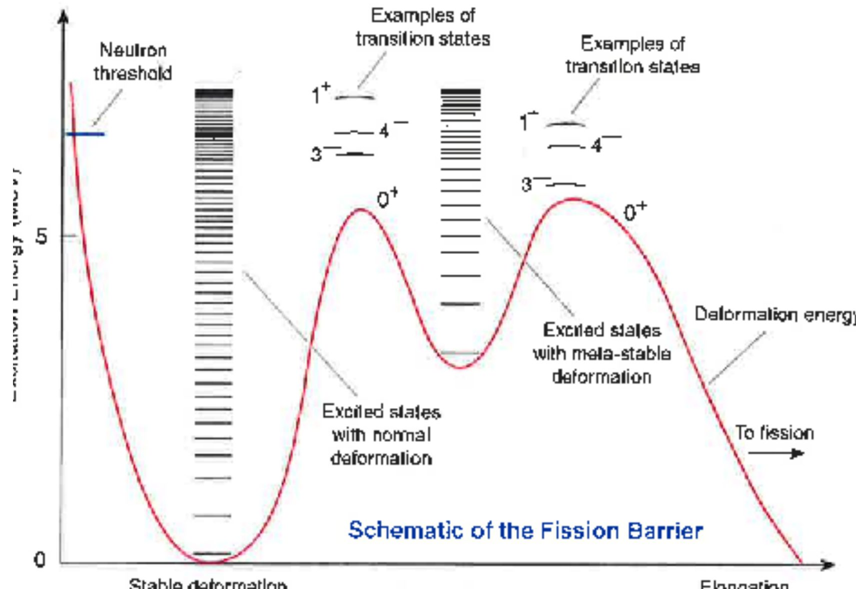
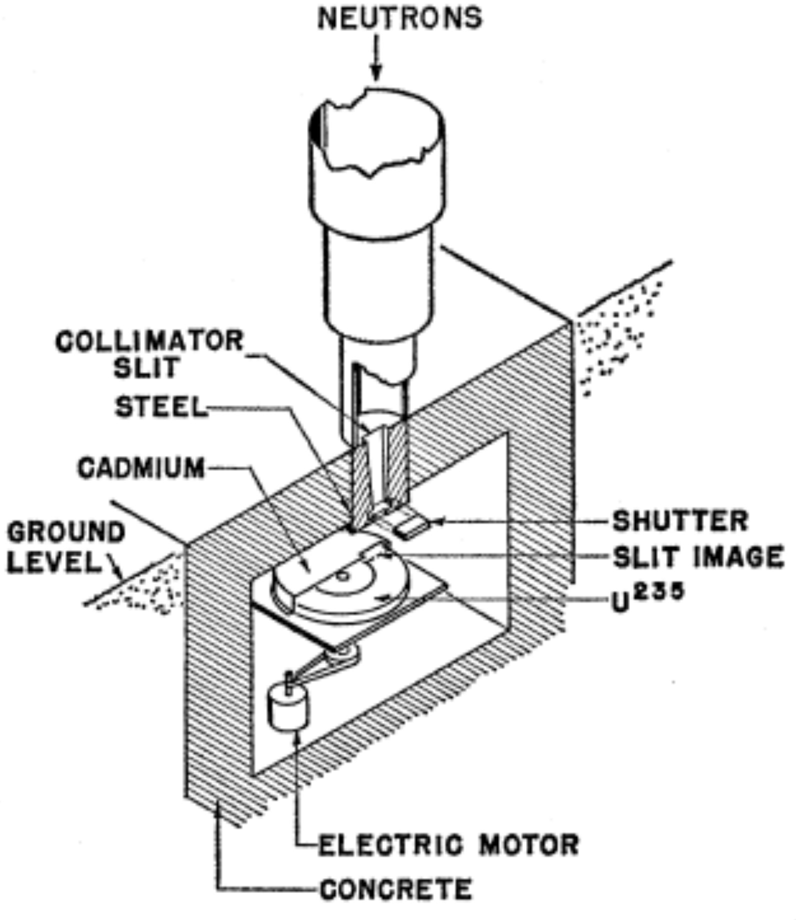
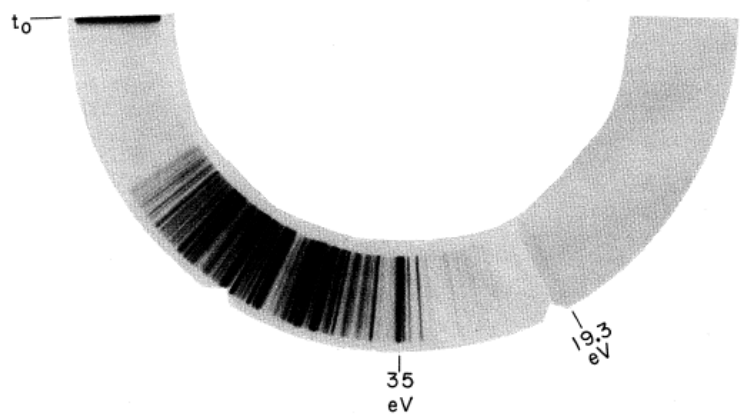


Figure 2.

The figure above represents the effect of compound nuclear spin on fission.

The Los Alamos Wheel Experiments (1958-1965)

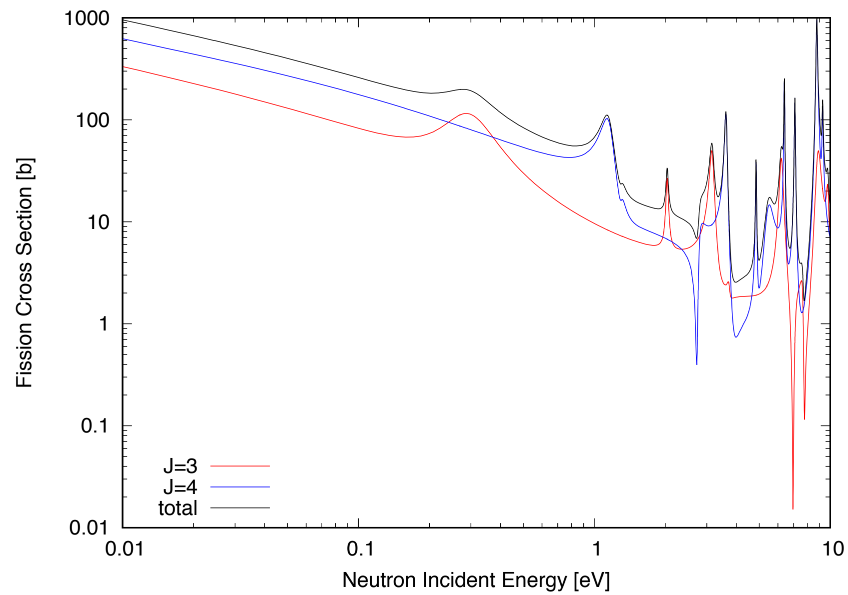




Neutrons were moderated at the source, peaking at 40-70 eV. The 2 foot diameter wheel rotated at 49 revolutions per second. The flight path was approximately 240 meters long.

Apply Results of the Wheel Experiment to ENDF

- The ENDF U-235 fission cross section can be separated into J=3-, and J=4- components.
- Resonances of the same angular momentum can interfere with each other under the Reich-Moore formalism.
- States of different angular momentum are orthogonal.
- Angular momentum and parity are conserved in the fission process.

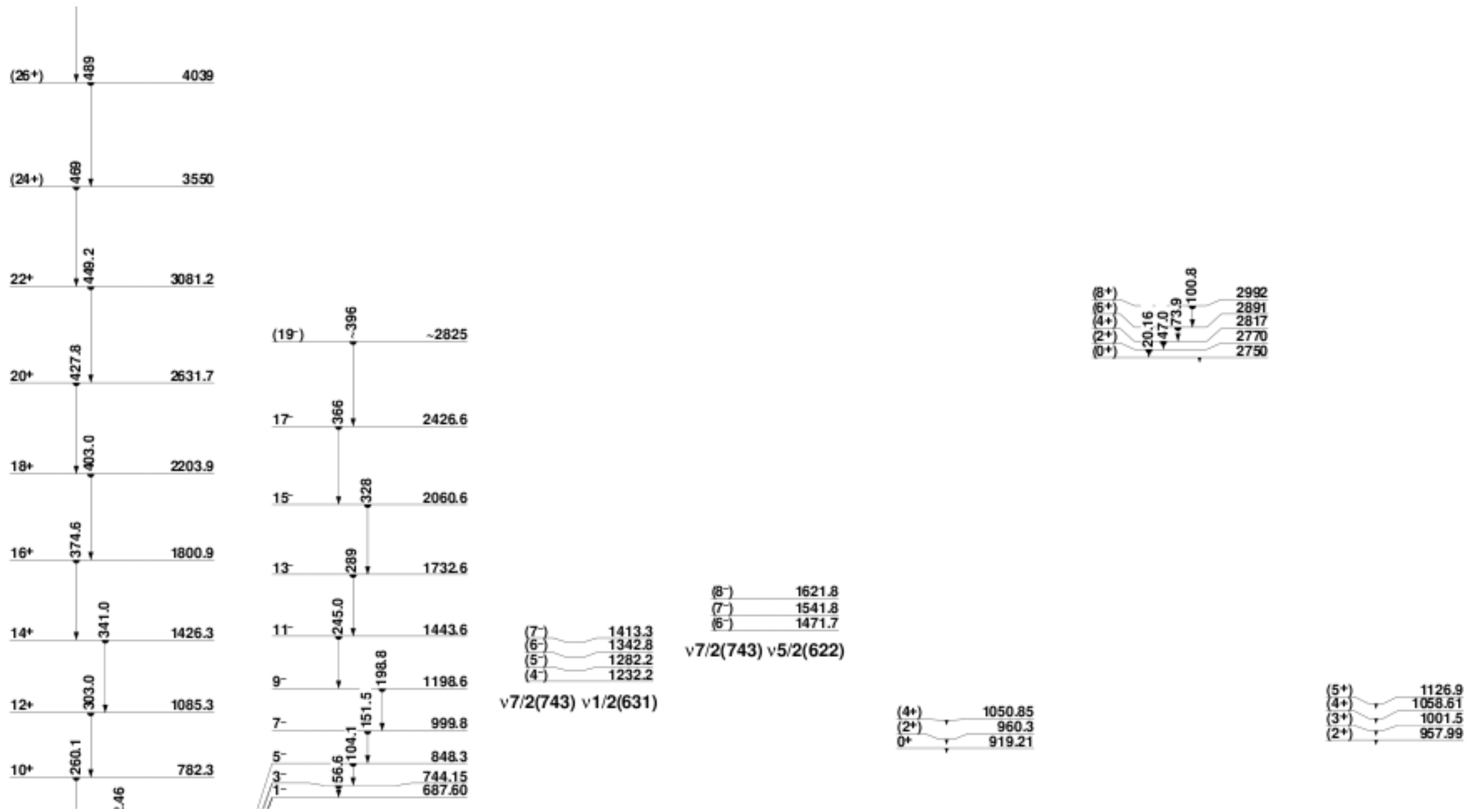


Application of the Wheel Results to ENDF

Spectrum	Contribution to fission cross section		R-111		R-115g	
	J=3-	J=4-	Group I	Group 2	Group I	Group 2
Thermal	21%	79%	0.69	1.08	0.593	1.11
Epithermal	38%	62%				

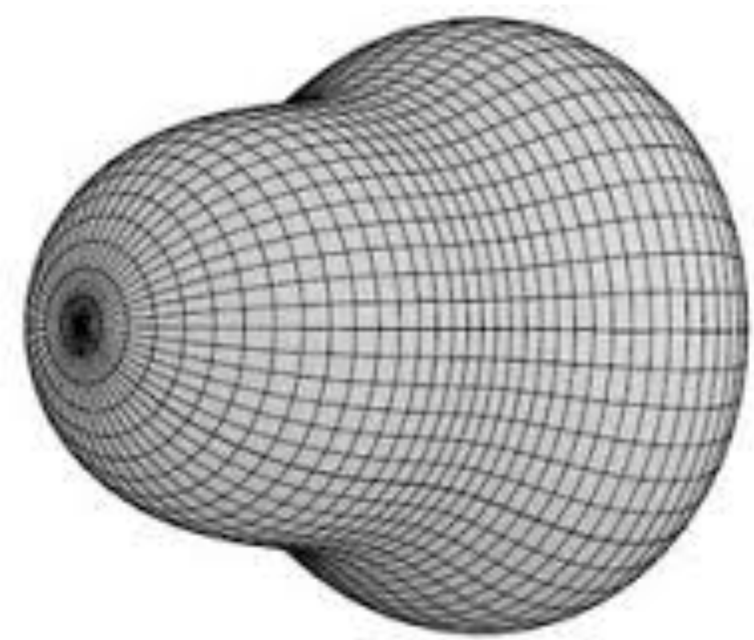
- The experimental values R-115g in a thermal neutron spectrum and an epithermal spectrum are, 1.00 and 0.89 ±0.02, respectively.
- If J=3- is assumed to correspond to group 2 (the more symmetric) the results for a thermal neutron spectrum and an epithermal spectrum are,0.70 and 0.79, respectively.
- If J=3- is assumed to correspond to group 1 (the more asymmetric) the results for a thermal neutron spectrum and an epithermal spectrum are,1.00 and 0.91, respectively.
- The experimental values R-111 in a thermal neutron spectrum and an epithermal spectrum are, 1.00 and 0.84 ±0.03, respectively.
- Likewise the R-111 results only agree if J=3- corresponds to the more asymmetric group.

Level Structure of U-236



Why the 3- Level Makes Sense as Leading to Asymmetric Fission

- The 3- level in U-236 belongs to an octupole band. The figure to the right illustrates an octupole deformation
- The 4- is base of a 2 quasi-particle K=4 band. The K band is stabilized by orienting is angular momentum vector along the axis of quadrupole deformation.
- This is an interesting observation of the effect of nuclear structure in the compound nucleus on the fission process.



Summary

- Radiochemistry has been used to study fission since it's discovery.
- Radiochemical measurement of fission product yields have provided the highest precision data for developing fission models and for nuclear forensics.
- The two-mode fission hypothesis provides a description of the neutron energy dependence of the mass yield curve. However, data is still rather sparse and more work is needed near second and third chance fission.
- Radiochemical measurements have provided evidence for the importance of nuclear states in the compound nucleus in predicting the mass yield curve in the resonance region

Acknowledgements

- Thanks to the nuclear and a radiochemistry group for R-value data.
- Thanks to Colorado School of Mines and the USGS Denver reactor for providing facilities.
- Thanks to Prof. Jenifer Braley and Dr. Michael Koehl for his doctoral thesis work.
- Thanks to the Los Alamos critical assembly group for neutron irradiations.
- Thanks to the NSTEC DPF group for neutron irradiations.
- This work was partially supported by NA-22 of the DOE-NNSA.

Backup Slides

Results of the Plutonium Wheel Experiment

E_0 (eV)	r-115	R-111	J
15.42	7.80E-03	13.4	1
17.66	5.00E-03	12.0	1
22.28	1.05E-02	14.7	1
26.29	7.50E-03	13.3	1
32.38	4.70E-02	32.5	0
41.52	6.70E-03	12.9	1
44.59	1.14E-02	15.2	1
47.74	6.90E-03	13.0	1
49.85	2.33E-02	21.0	0
50.22	7.00E-03	13.0	1
52.74	1.03E-02	14.6	1
55.79	4.00E-03	11.5	1
57.6	2.83E-02	23.4	0
58	2.83E-02	23.4	0
59.39	1.36E-02	16.2	1
61.1	3.02E-02	24.3	0
65.96	1.94E-02	19.1	1
66.83	2.79E-02	23.2	0
75.21	6.80E-03	12.9	1
81.7	2.85E-02	23.5	0

- ***The average R-111 for J=0 and J=1 resonances are 24.5 and 14.0, respectively.***
- ***The thermal R-111 is 16.5.***
- ***We infer that at thermal neutron energy the fission cross section is 76 percent J=1 and 24 percent J=0.***
- ***The resonances above threshold up to 14.6 eV are J=1. The implied contribution from a J=0 bound state is about 24 percent.***

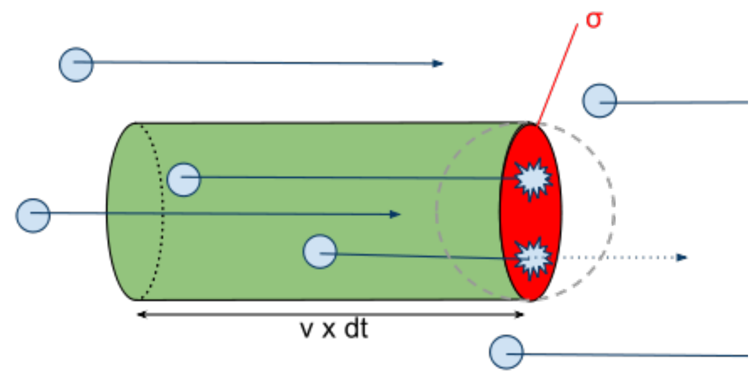
Results of the U-235 Wheel Experiment

E-zero	R-115	R-111	J
19.3	4.02E-01	0.59	4
21.07	5.17E-01	0.69	4
22.94	3.74E-01	0.57	4
23.63	3.28E-01	0.53	4
24.23	1.10E+00	1.17	3
25.65	1.03E+00	1.11	3
26.44	6.19E-01	0.77	4
27.81	5.54E-01	0.72	4
28.35	1.25E+00	1.29	3
30.91	4.41E-01	0.62	4
32.1	7.08E-01	0.84	4
33.55	4.01E-01	0.59	4
34.39	1.04E+00	1.11	3
34.85	8.30E-01	0.94	4
35.21	1.03E+00	1.11	3
35.75	1.33E+00	1.35	3
36.64	1.36E+00	1.38	3
38.42	1.04E+00	1.11	3
39.44	1.01E+00	1.09	3
40.51	6.75E-01	0.82	4
41.91	6.95E-01	0.83	4
43.41	5.44E-01	0.71	4
44.04	6.33E-01	0.78	4
44.75	1.06E+00	1.13	3
45.79	5.74E-01	0.73	4
46.93	6.20E-01	0.77	4
48.06	7.69E-01	0.89	4
48.82	8.75E-01	0.98	4
49.51	9.67E-01	1.06	3

- ***Average R-111 was determined from 24 resonances with J=4 and 14 resonances with J=3.***
- ***The average R-111 for J=3 and J=4 resonances are 1.51 and 0.75, respectively.***
- ***The thermal R-111 is 1.0.***
- ***We infer that at thermal neutron energy the fission cross section is 67 percent J=4 and 33 percent J=3.***
- ***The resonances above threshold are J=3. A resonance at threshold plus bound states contribute about 67 percent to J=4.***

Basic Concepts: Nuclear Cross Section

- The reaction rate is equal to the number of target atoms times the nuclear cross section, times the neutron flux.
- The flux, ϕ, has units of neutron per square centimeter per second. It is sometimes given as neutron density times neutron velocity.
- The cross section, σ, is given in the unit barn, 1×10^{-24} cm^2.
- The integrated flux, ϕt, is called the fluence.



$$\frac{dN_{A+1}}{dt} = N_A\sigma\phi = N_A\sigma nv$$

$$N_{A+1} = \int_0^t N_A\sigma\phi dt$$

$$= N_A\sigma\phi t$$

Basic Concepts: Level Widths

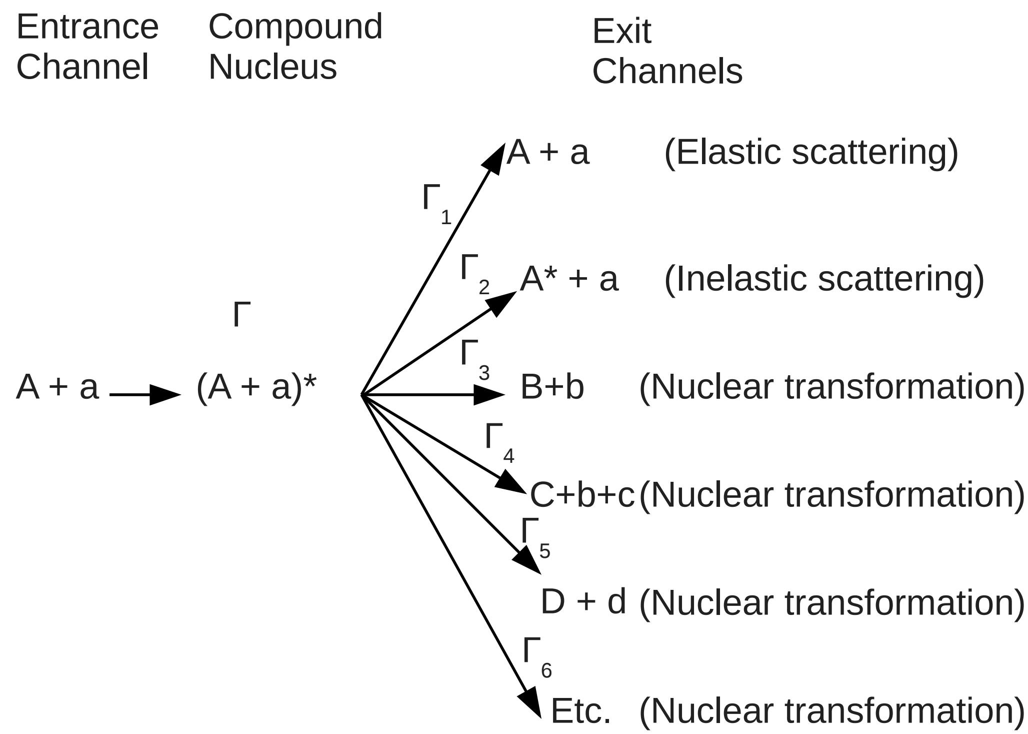
- The relationship between the excited state level width and lifetime comes from the time-dependent Schrödinger equation.
- The half-life is ln(2) times the life-time.

$$\hbar = \Delta E \Delta t$$

$$\tau = \frac{\hbar}{\Gamma}$$

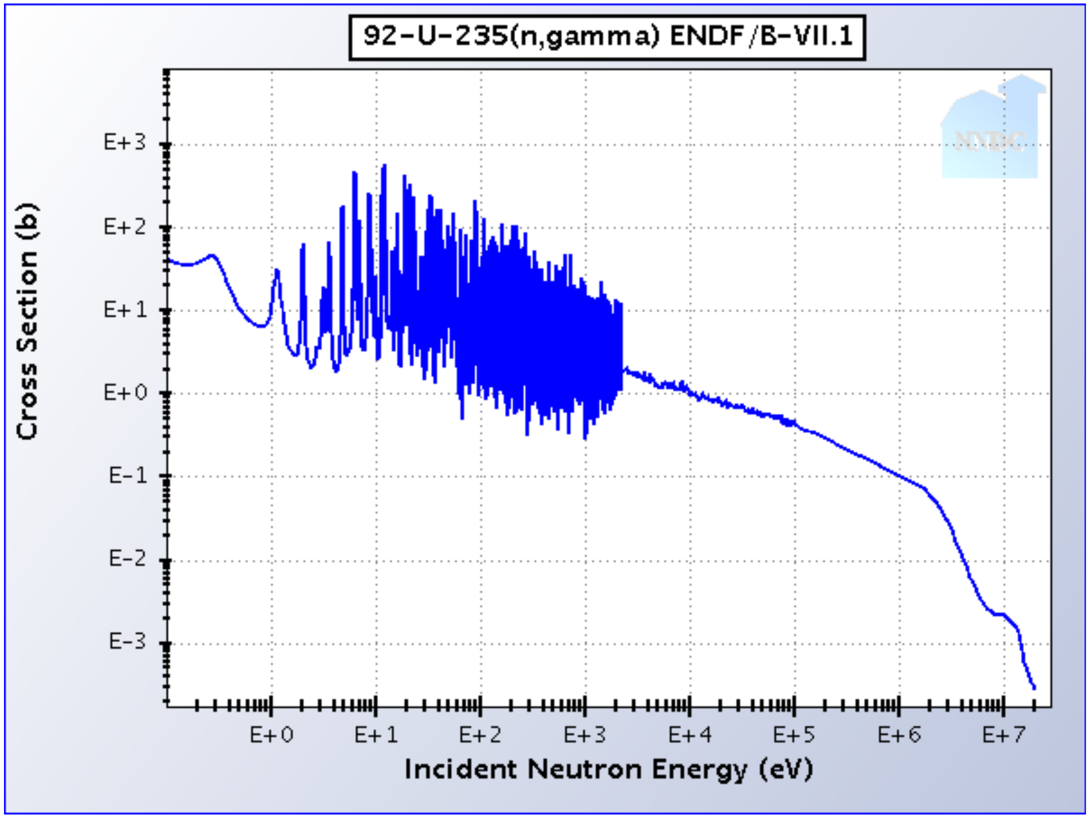
Basic Concepts: The Compound Nucleus

- **Niels Bohr developed the concept of the compound nucleus.**
- **A particle, e.g., neutron, enters the nucleus creating a new nucleus in an excited state.**
- **The life-time of this state is determined by the total width of the level.**
- **The total width is the sum of the partial widths of the possible exit channels.**
- **The partial width over the total width can be thought of as the probability of that exit channel.**



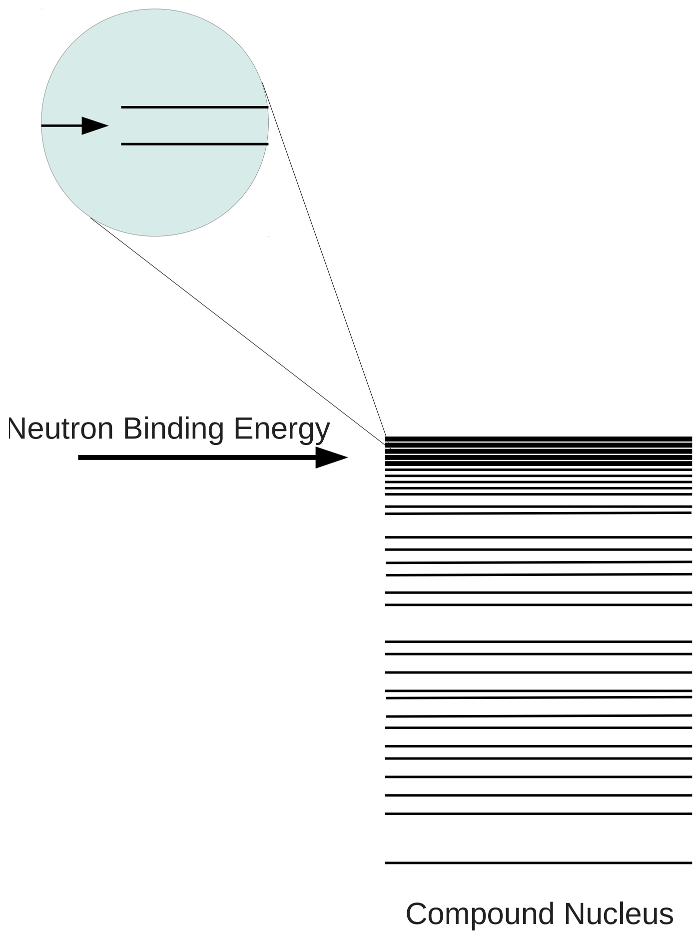
Basic Concepts: Neutron Capture

- **Neutron capture has no barrier to overcome.**
- **The compound nucleus decays primarily by gamma ray emission.**
- **Resonances can be observed at low neutron kinetic energy.**
- **Resonances can't be resolved at neutron energies much above 2 keV.**



Basic Concepts: Levels in the Compound Nucleus

- The neutron binding energy corresponds to zero neutron kinetic energy.
- When the neutron kinetic energy plus binding energy equals an excited state the probability for capture is greatly enhanced. This is analogous to optical spectroscopy.
- Neutron capture can proceed by gamma ray emission.
- Other channels are fission, elastic, and inelastic scattering.



Basic Concepts: The Breit-Wigner Formula

$$\sigma(n,\gamma) = \pi\lambda\!\!\!^{-2}\left[\frac{2I_c+1}{(2I_n+1)(2I_t+1)}\right]\frac{\Gamma_n\Gamma_\gamma}{(E_n-E_0)^2+(\Gamma/2)^2}$$

$$\Gamma_n = \Gamma_n^o\sqrt{\frac{E_n}{1eV}}$$

$$\sigma(n,\gamma) \propto 1/v$$

- **The cross section is proportional to the de Broglie wavelength squared.**
- **There is a statistical spin factor, the spin multiplicity of the compound nucleus, over the spin multiplicity of the neutron (2) times the spin multiplicity of the target.**
- **The shape of the resonance is described by a Lorentzian function, where the full width half maximum is the total width.**
- **At high energy 1 MeV and above, the de Broglie wavelength is much smaller than the nucleus. The total nuclear cross section becomes the geometric cross section.**

Plutonium-239 Fission Yield of Neodymium-147

- Nd-147 is an important fission product in nuclear weapons diagnostics.
- It has the same volatility as plutonium.
- It is assumed that the yield does not depend on neutron energy in the vicinity of a fission spectrum.
- Recent evaluations suggest that this assumption may be incorrect.
- The fission yield could be 5 percent higher at fission energies than thermal.
- D-D neutrons from the DPF have an energy of 2.9 MeV due to beam-target interactions.
- 2.9 MeV has a good lever arm for measuring the energy dependence.
- The 14 MeV measurements of yield scatter by as much as 40 percent.